UDC 666.3-127

MULTILAYER POROUS ALUMINOSILICATE CERAMIC

S. M. Azarov,[1] T. A. Azarova,[1] A. I. Rat'ko,[1] N. V. Mil'vit,[1] and S. V. Shemchenok[1]

Translated from *Steklo i Keramika*, No. 3, pp. 23 – 25, March, 2008.

Multilayer aluminosilicate materials used for treating drinking water were investigated. The basic results of the experimental studies obtained in developing the process conditions for fabrication of aluminosilicate filter elements are reported.

Porous ceramics are widely used for technologies for treating liquids and gases. Ceramic filters are used for mechanical and biological treatment of water, treatment of liquids in the food and pharmaceutical industry, and for capturing valuable and toxic components in gaseous systems.

Most multilayer filter ceramics are molded according to the following principle: one or more filtering layers of fixed porosity are applied on a macroporous support. The macroporous base provides for mechanical strength and has high permeability in sufficiently intensive flow of the filtered medium [1, 2].

Since application of the filtering (membrane) layer directly on a large-pore base is not always effective, an intermediate layer (mesolayer) is formed first. It can reduce clogging of the selective layer by small particles and serve as its support. The mesolayer should smooth the internal pressure differential in the membrane layer, reducing the filtered medium flow resistance.

The membrane layer is responsible for the selectivity of the process and limits its productivity.

An analysis of the published data shows the promise of using multilayer porous ceramic materials for production of filter elements based on a set of output – treatment quality characteristics. At the same time, the problem of increasing the work life of filter elements while preserving output and porosity is becoming acute. We investigated the effect of the conditions of molding the filter layers on the performance characteristics of multilayer aluminosilicate filter elements.

The technology for manufacturing multilayer filter elements from aluminosilicate powder consisted of the following operations: mixing the initial components, molding and sintering the stock for the bases of the filter elements, fabrication of slip, and application of filtering (intermediate and membrane) layers followed by sintering.

[1] Institute of General and Inorganic Chemistry, National Academy of Sciences of Belarus, Minsk, Belarus.

Aluminosilicate powder, kaolin raw material, and a blowing agent were used as the initial components for fabricating the bases of the filter elements.

The large-pore bases of the filter elements were molded in a radial molding unit that ensured uniform compression of the stock over the entire length [3]; sintering was conducted in an electric furnace for sintering ceramic materials at a temperature of 1150°C. Aluminosilicate powder separated into 10 – 50, 50 – 100, 50 – 200, and 100 – 200 μm fractions was used for application of the filtering layers. The membrane layer was molded with the same powder with a particle size of less than 10 μm. The studies were conducted on cylindrical samples with an external diameter of 45 mm, height of 53 – 56 mm, and wall thickness of 4 – 5 mm. The open porosity of the materials was determined with the method reported in [1], and the pore size corresponded to GOST 26849–86.

The lifetime of the filter elements (before they became totally clogged with filtration products) was tested in a standard frame. After removal from the frame, the used, previously dried samples were weighed. The difference in the weight of the unit of volume of the samples before and after the tests related to the unit of filtering surface area was used as the degree of contamination of the filter elements.

The output of the filter element, $m^3/(m^2 \cdot sec)$, was calculated with the equation:

$$v = \frac{V}{St},$$

where V is the amount of water passing through the filter element, liters; in the experimental conditions, $V = 5 \times 10^{-3}$ m^3; S is the area of the external surface of the filter element, m^2; t is the time in which the water passed through the filter element, sec.

TABLE 1

Index	After heat treatment	
	first	second
Open porosity, %	22.7	22.8
Pore size, μm:		
maximum	51.3	56.8
average	27.1	27.0
Output, 10^{-3} $m^3/(m^2 \cdot sec)$	34.1	40.0

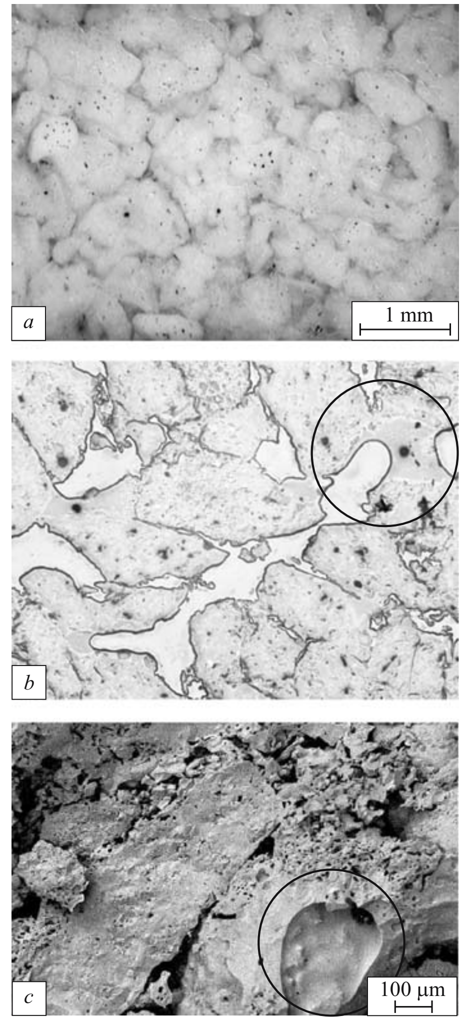

Fig. 1. Structure of the material in the base of multilayer aluminosilicate ceramics: *a*) surface (optical microscope); *b*) fracture (microsection, optical microscope); *c*) fracture (electron microscope).

The structure of the filter element materials was investigated in Neophot 32 and JEOL JSM-840 microscopes. The mass concentration of total iron in the drinking water was measured before and after filtration by the photocolorimetric method in GOST 4011–72.

The material of the base of the multilayer aluminosilicate ceramic had an alveolar structure [4] with a slit-like pore structure (Fig. 1). According to the data from x-ray phase analysis, it consisted of mullite, quartz, and cristobalite.

In molding the filtering layers, the material of the porous base underwent repeated high-temperature treatment. As the studies showed, some of its characteristics changed. The averaged results of the comparative studies of the characteristics of the multilayer material bases before and after the second heat treatment at the temperature of molding the intermediate layer are reported in Table 1.

As the results suggest, repeated sintering was accompanied by an increase in the output of the base to 17%. The open porosity and average pore size of the samples remained almost unchanged. These results can be explained by considering the fact that the filter element base material was molded in the presence of a liquid phase, whose formation was confirmed by the presence of the characteristic structural fragments shown in Fig. 1*b* and c. In repeated sintering, the structural transformations in the aluminosilicate material took place by bulk redistribution of a low-melting constituent. The output (throughput) of the sample did not increase due to a change in the porosity of the material but due to the formation of large pores, which was confirmed by the results of measurements of the maximum pore size in the filter element bases.

The comparative characteristics of the elements whose filtering layers were molded with aluminosilicate powder of different fractional composition and the results of performance tests to determine the quality of the treated water, output, and sediment capacity are reported in Table 2.

In analyzing the results obtained, we can conclude that the formation of layers on the surface of the porous aluminosilicate base by powders of different fractional composition allow fabricating filter elements with different performance characteristics. The finer the powder used, the smaller the filter element pore size, the longer the operating life, and the higher the degree of treatment of the water with respect to iron (see Table 2, elements 2, 4, and 5). In molding the layers, it is desirable to use powders from a narrow fraction, since important difference sin the particle size since the large pores in the filtering layer material formed by large particles are clogged by the fine fraction of the powder. This is the cause of the reduced operating life of the filtering layer made of powder of the widest fractional composition (50 – 200 μm), which is not as much due to the small average pore size as to the absence of regularity of the structure of the pore space, which perturbs the permeability of the material. For this reason, the filter material has minimal sediment capacity.

The character of the change in the output of aluminosilicate filter elements with filtering layers made of powders of different fractions in time is shown in Fig. 2. As the studies showed, for all of the filter elements, the basic loss of output in filtration of water takes place almost simultaneously after the first 0.25 m^3 and is not a function of the initial value.

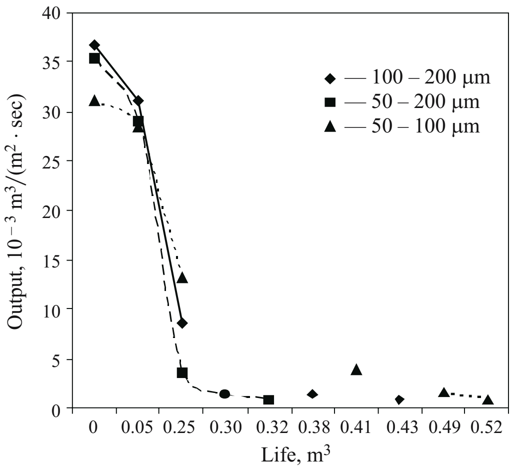

Fig. 2. Dynamics of loss of output of samples of aluminosilicate filter elements with filtering layers made of powders from different fractions.

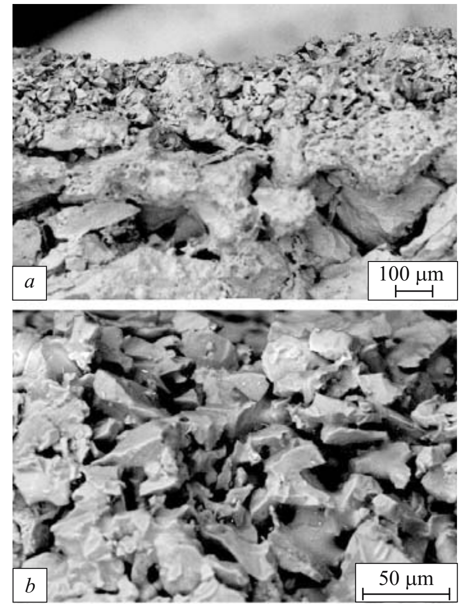

Fig. 3. Structure of a multilayer filter element with a filtering layer made of powder from the 10 – 50 μm fraction (two-stage application; scanning electron microscope): *a*) fracture in radial direction; *b*) filtering layer (viewed from above).

As a result of many experiments, it was found that increasing the thickness of the selective layer of the filter element (due to the number of layers applied) is only justified for powder of the 10 – 50 μm fraction, and then only when it is necessary to significantly increase the degree of treatment of the water to remove iron. Five applications of filtering layers with 10 – 50 μm particle size in comparison to two applications reduced the iron content in the water by almost 3 times (see Table 2, elements 5 and 7). However, such filter elements have much worse output and sediment capacity than filter elements with a double layer.

Filter elements with two and three layers made of 10 – 50 μm powder had the best performance characteristics. However, since the two-layer filter element has maximum sediment capacity but is insignificantly worse than the three-layer element in output and water treatment quality while the production technology is less laborious, it is the most promising. The structure of the material and its filtering layer are shown in Fig. 3.

Attempts to improve the performance characteristics of the two-layer element with a filtering layer made of 10 – 50 μm powder by application of intermediate layers of larger fractions did not produce the expected positive results. Application of a membrane layer (see Table 2, elements 5 and 8) allows insignificantly improving the quality of treatment of the water with respect to iron, but significantly decreases the sediment capacity of the filter element.

TABLE 2

Element	Fractional composition of applied layer powder, μm	Number of layers	Operating life, m^3	Sediment capacity, g/m^2	Iron content in water after filtration, mg/liter	Pore size, μm	
						maximum	average
1	Filter element base	0	Almost unlimited	0	Initial	56.8*	27.0*
2	100 – 200	2	0.43	12.5	0.0339	44.9	22.0
3	50 – 200	2	0.32	6.3	0.0405	48.6	16.9
4	50 – 100	2	0.52	12.4	0.0314	50.3	17.9
5	10 – 50	2	0.83	74.7	0.0290	42.4	16.3
6	10 – 50	3	1.04	24.6	0.0222	45.6	17.2
7	10 – 50	5	0.31	12.6	0.0101	40.1	16.5
8	10 – 50**	2	0.71	12.3	0.0240	33.7	14.0

* For correctness of the experiment, samples of the bases underwent additional heat treatment based on the molding conditions for the filtering layer.

** With a membrane layer formed with aluminosilicate particles less than 10 μm in size.

Based on the data from the studies, the filter elements do not affect the pH and do not change the hardness of the water.

Filter elements with two and three layers made of 10 – 50 μm powder thus have the best performance characteristics. The filter elements have output of (30.6 – 37.5) × 10^{-3} $m^3/(m^2 \cdot sec)$ as a function of the dispersion of the particles forming the layer on a large-pore aluminosilicate support.

The operating life of the filter element can be significantly increased by installing on its surface a fabric filter made of polyester filter cloth which plays the role of a preliminary filtering layer.

REFERENCES

1. K. A. Smirnova, *Porous Ceramic for Filtration and Aeration* [in Russian], Stroiizdat, Moscow (1968).
2. V. N. Pavlikov and E. P. Garmash, *Ceramic Membranes* [in Russian], Naukova Dumka, Kiev (1992).
3. O. P. Reut, L. S. Boginskii, and E. E. Petyushik, “Radial molding of condensed materials. Scheme, equipment, technology, articles,” in: *Proc. of the International Scientific and Technical Conference on Powder Metallurgy: Advances and Problems* [in Russian], Minsk (2005), pp. 90 – 93.
4. I. Ya. Guzman, “Some principles of formation of porous ceramic structures. Properties and use,” *Steklo Keram.*, No. 9, 28 – 31 (2003).